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SELENIUM SEVENTY-FIVE

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ABSTRACT

Evidence has been given to show that the 115 day activity produced in selenium by thermal neutrons is due to Se^{75} which decays by K-electron capture to As^{75}, accompanied by a 0.4 Mev γ ray. A value for the cross section of Se^{74} for radiative capture of a thermal neutron has been given.

*   *   *   *   *

Seaborg's tables[1] report as a K-capture process a 48 day and a 160 day activity for Se^{75} produced from As^{75} by a (p,n) and (d,2n) reaction respectively. However we found a K-capture activity with a half-life of 115 ± 5 days produced by a (n,γ) reaction on selenium irradiated in the Argonne Pile. The remainder of this paper will give evidence for this activity and for a value of the activation cross section for its production from Se^{74}.

The 115 day activity was established by counting samples mounted on scotch tape foils with an aluminum walled cylindrical Geiger counter. One sample was counted for over six months.

Possibility of an arsenic activity produced by a (n,p) reaction on the selenium was ruled out by chemical separations from the selenium using an arsenic carrier. No activity was found in the arsenic fractions. The chemical separation was checked by adding 27 hour As^{76} to nonactivated selenium and separating the two elements. No arsenic activity was found in the selenium portions. Similar separations with germanium ruled out the possibility of a (n,α) reaction on the selenium to produce germanium. The possibility of the activity being due to impurities in the selenium was ruled out by using a sample of Hilger spectroscopically standardized selenium reported to be 99.9964% pure. Samples of selenium of doubtful purity showed this activity to be fully as strong as the Hilger sample. The only other elements exhibiting half-lives of this order of magnitude and having large cross sections and which could be impurities (since they have similar chemical properties with selenium) were tellurium and tantallum. Both of these were eliminated by spectroscopic analysis of the Hilger sample.

It then remained to determine the selenium isotope responsible. Since a possible reaction was a (n,γ) reaction on Se^{82} to produce Se^{83} with subsequent beta decay to Br^{83} which has a 140 minute half-life, a search for a bromine daughter was undertaken. A bromide ion carrier was added to a selenium solution in hydrochloric acid. After allowing 24 hours for the possible bromine daughter to grow, bromide ion was separated from selenium ion. He activity was found in the bromine fraction. Thus eliminating Se^{83}. The only other possibilities were Se^{75}, Se^{79}, and Se^{81}. The two latter would be negative beta emitters whereas the first would be a positron emitter or produce arsenic K-x-rays by K-capture. Cloud chamber studies showed that no beta particles (positive or negative) were emitted as will be discussed below. Further evidence for Se^{75} was obtained by bombarding arsenic, which consists of only the one stable isotope As^{75}, with 12 Mev deutrons from the cyclotron at Washington University in St. Louis. The only possible selenium isotope which could be produced (this by a (d,2n) reaction) for which no stable isotope was known was Se^{75}. The selenium thus produced was then separated from the arsenic. The decay of the sample also gave a 115 ± 5 day half-life and the mass absorption coefficients for this sample were the same as those for the sample produced by the pile neutrons on selenium.

A study of the radiation emitted showed a γ ray of about 0.4 Mev as determined by an absorption curve taken with lead absorbers after electrons and x-rays were absorbed by one centimeter of lucite. An absorption curve taken on the main radiation with aluminum absorbers gave a mass absorption coefficient of 42 cm^2/g. This is roughly the value to be expected for x-rays from elements like arsenic or selenium. If K-electron capture were the mode of decay of Se^{75}, arsenic K-x-rays would be expected. However, if positron emission were the mode of decay, any x-rays accompanying them would be selenium x-rays. Table 1[2] gives the values of the K critical absorption wavelengths for several absorbers. X-rays of wavelengths slightly shorter than the K critical values for an absorber are strongly absorbed by it. Table 2[3] gives the main K-x-ray lines for arsenic, selenium, and germanium. We see that if selenium x-rays are present they would be more strongly absorbed by germanium or gallium absorbers than by arsenic absorbers. Whereas arsenic x-rays would be more strongly absorbed in gallium or zinc absorbers than germanium absorbers. While if germanium x-rays are present they would be more strongly absorbed in zinc absorbers than gallium absorbers. Table 3 gives the values of the mass absorption coefficient as measured by us for various absorbers. Since gallium shows stronger absorption than germanium we must conclude that arsenic x-rays are present due to K-capture in Se^{75}.

Table 1.   Table 2.

Element	K-absorption edge	Element	$K-\alpha_1$	$K-\alpha_2$
Zn	1.2805 A	$_{34}$Se	1.10248 A	1.10652 A
Ga	1.1902 A	$_{33}$As	1.17344 A	1.17743 A
Ge	1.1164 A	$_{32}$Ge	1.25130 A	1.25521 A
As	1.0426 A			

Table 3.

Absorber μ/ρ	in cm^2/g
$_{13}$Al	42
$_{30}$Zn	115
$_{31}$Ga	102
$_{32}$Ge	56
$_{33}$As	56

In order to completely verify the presence of x-rays and the absence of β - particle decay, a foil of the active selenium was placed in a cloud chamber.* With argon gas in the chamber many short photoelectrons, of range about one millimeter, originating in the gas were observed, but no longer electrons (above the background) originated at the foil. With helium gas in the chamber only a few photoelectrons were seen in the gas and an upper limit on the number of electrons starting from the foil could be fixed. These results showed that less than 1% of the disintegrations could be by positron emission whereas the major portion is by K-electron capture with subsequent emission of As K-x-rays.

The activation cross section[4] for the production of Se^{75} from Se^{74} by a thermal neutron (n,γ) process was calculated on the basis of the cloud chamber tracks, on an estimation of the counter efficiency for the arsenic K-x-rays, and on an estimation of the counter efficiency for the γ rays produced. The calculations by the three methods checked fairly well and a weighted average is reported here: $22 \times 10^{-24} cm^2$/isotopic atom or 0.2×10^{-24} cm^2/natural atom since the natural abundance of Se^{74} is 0.9%. The probable error is estimated to be about $\pm 40\%$.

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*The cloud chamber work was done by D. Hughes and C. Egler.

The chemical procedures of separation used were the following: A solution of selenium was prepared by dissolving selenium metal powder in concentrated nitric acid, evaporating to dryness, and redissolving in a minimum of concentrated HCl. Arsenous oxide or germanium oxide carrier was then added and the selenium precipitated by reduction to the metal with sulfur dioxide. The precipitated metal was washed with water and then ethyl alcohol and dried at 110°C. The arsenic of germanium was then precipitated from the acid solution as a sulfide by hydrogen sulfide.

In the case of the cyclotron bombardment, the arsenous oxide was dissolved in concentrated HCl, selenous oxide carrier was added and the selenium was precipitated by reduction with sulfur dioxide.

To look for the possible bromine daughter, selenium was dissolved and the bromine was allowed to grow for 24 hours in the presence of added sodium bromide carrier. Then silver nitrate was added precipitating silver selenide and bromide which were then redissolved in concentrated aqueous ammonia. The silver bromide was then reprecipitated from hot nitric acid solution in which silver selenide remains soluble.

The mass absorption coefficients were obtained by placing foils of known thickness of the suitable element between the sample and a bell type, thin mica window Geiger counter. The absorbers of gallium, germanium, and arsenic were prepared by placing known amounts of the powdered oxides on scotch tape.

REFERENCES

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3. Ibid., page 784.

4. Hughes, D. and C. Egler, For a fuller discussion of cross section
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